

TABLE 2

Solid electrolyte																
A							B									
Test No	No.	Type	Manu-fac-turing method	Pa $\mu$ m 2-0.4	Pa90 $\mu$ m 3.4-0.7	WPa parts by mass	No.	Type	Manu-fac-turing method	Pb $\mu$ m 1.5-0.1	Pb90 $\mu$ m 2.5-0.2	WPb parts by mass	Pb/Pa 0.05-0.75	WPb/ (WPa + WPb) 0.01-0.8	Porosity	Ion conduc-tance
201	PSI	Sulfide	Wet	1.5	2.5	100	PS2	Sulfide	Wet	0.9	1.5	20	0.60	0.17	A	A
c21	PSI	Sulfide	Wet	1.5	9.5	100	—	—	—	—	—	—	—	—	C	C

[0210] Sulfide: A sulfide inorganic solid electrolyte (Li/P/S-based glass) synthesized as below

[0211] Synthesis of a Sulfide Inorganic Solid Electrolyte (Li/P/S-Based Glass)

[0212] 2.42 g of lithium sulfide ( $\text{Li}_2\text{S}$ , manufactured by Sigma-Aldrich Co., LLC., purity>99.98%), and 3.90 g of diphosphorus pentasulfide ( $\text{P}_2\text{S}_5$ , manufactured by Sigma-Aldrich Co., LLC., purity>99%) were respectively weighed in a glove box under argon atmosphere (dew point:  $-70^\circ\text{C}$ .), and were introduced to a mortar.  $\text{Li}_2\text{S}$  and  $\text{P}_2\text{S}_5$  satisfied  $\text{Li}_2\text{S}:\text{P}_2\text{S}_5=75:25$  in the molar ratio. In the agate mortar, mixture was performed for five minutes by using agate pestle.

[0213] 66 zirconia beads having the diameter of 5 mm were introduced to a 45 mL zirconia container (manufactured by Fritsch Japan Co., Ltd.), the total amounts of the mixture described above were introduced, and the container was completely sealed under argon atmosphere. The container was set to a planet ball mill P-7 manufactured by Fritsch Japan Co., Ltd., and 6.20 g of a yellow powder sulfide solid electrolyte material (Li/P/S glass) was obtained by performing mechanical milling at  $25^\circ\text{C}$ . and the number of rotations of 510 rpm for 20 hours.

[0214] Subsequently, 160 zirconia beads having the diameter of 5 mm were introduced to a zirconia 45 mL container (manufactured by Fritsch Japan Co., Ltd.), 9.0 g of an sulfide inorganic solid electrolyte (Li/P/S glass), 0.3 g of HSBR (DYNARON 1321P manufactured by JSR Corporation) as a binding material, and 15.0 g of toluene as a dispersion medium were added, the container was set to a planetary ball mill P-7 manufactured by Fritsch Japan Co., Ltd., and the wet dispersion was performed for 90 minutes at the rotation speed of 360 rpm, so as to obtain sulfide solid electrolyte particles PS1. The average particle diameter was 1.5  $\mu\text{m}$ , and the accumulative 90% particle diameter was 2.5  $\mu\text{m}$ .

[0215] Separately, 160 zirconia beads having the diameter of 5 mm were introduced to a zirconia 45 mL container (manufactured by Fritsch Japan Co., Ltd.), 9.0 g of an sulfide inorganic solid electrolyte (Li/P/S glass), 0.3 g of HSBR (DYNARON 1321P manufactured by JSR Corporation) as a binding material, and 15.0 g of toluene as a dispersion medium were added, the container was set to a planetary ball mill P-7 manufactured by Fritsch Japan Co., Ltd., and the wet dispersion was performed for 120 minutes at the rotation speed of 360 rpm, so as to obtain sulfide solid electrolyte particles PS2. The average particle diameter was 0.9  $\mu\text{m}$ , and the accumulative 90% particle diameter was 1.5  $\mu\text{m}$ .

[0216] The invention is described with reference to specific embodiments and drawings, but, unless described otherwise, it is clear that any details of the invention which are not particularly designated are not intended to limit the

invention, and it is obvious that the embodiments are widely construed without departing from the spirit and the scope of the invention recited in the accompanying claims.

#### EXPLANATION OF REFERENCES

- [0217] 1: negative electrode collector
- [0218] 2: negative electrode active substance layer
- [0219] 3: inorganic solid electrolyte layer
- [0220] 4: positive electrode active substance layer
- [0221] 5: positive electrode collector
- [0222] 6: operating position
- [0223] 10: all-solid-state secondary battery

What is claimed is:

1. A solid electrolyte composition comprising: inorganic solid electrolyte particles exhibiting at least two peaks in accumulative particle size distribution which is measured with a dynamic light scattering-type particle diameter distribution measuring device.
2. The solid electrolyte composition according to claim 1, wherein, among the two or more peaks, a peak (Pa) of a maximum particle diameter is in the particle diameter range of 2  $\mu\text{m}$  to 0.4  $\mu\text{m}$  and a peak (Pb) of a minimum particle diameter is in the range of 1.5  $\mu\text{m}$  to 0.1  $\mu\text{m}$ , and a relationship between the peak (Pa) of the maximum particle diameter and the peak (Pb) of the minimum particle diameter satisfies Expression (1) below.

$$0.05 \leq \text{Pb}/\text{Pa} \leq 0.75 \quad (1)$$

3. The solid electrolyte composition according to claim 1, wherein the inorganic solid electrolyte particles include inorganic solid electrolyte particles A having an average particle diameter (da) of 2  $\mu\text{m}$  to 0.4  $\mu\text{m}$  and inorganic solid electrolyte particles B having an average particle diameter (db) of 1.5  $\mu\text{m}$  to 0.1  $\mu\text{m}$ , and Expression (2) below is satisfied.

$$0.05 \leq \text{db}/\text{da} \leq 0.75 \quad (2)$$

4. The solid electrolyte composition according to claim 1, wherein, with respect to the accumulative particle size distribution measured with the dynamic light scattering-type particle diameter distribution measuring device, when respective peaks are assumed to follow log-normal distribution and the waveform is separated by a nonlinear least square method, an accumulative 90% particle diameter (Pa90) of a peak (Pa) of a maximum particle diameter is 3.4  $\mu\text{m}$  to 0.7  $\mu\text{m}$ , and an accumulative 90% particle diameter (Pb90) of a peak (Pb) of a minimum particle diameter is 2.5  $\mu\text{m}$  to 0.2  $\mu\text{m}$ .